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(54) Title: SOFT MAGNETIC ANISOTROPIC COMPOSITE MATERIALS

(57) Abstract

The invention concerns a soft magnetic, anisotropic composite material essentially consisting of compacted, essentially flaky shaped, electrically insulated particles of essentially pure iron powder containing less than 0.01 % by weight of carbon, which particles are aligned in essentially parallel relationship and bonded together by an organic polymer resin. The new material is characterized by a green density of at least 7.4 g/cm^3. The invention also concerns a process for the preparation of the new material as well as the use of the material for devices operating at power frequencies.

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SOFT MAGNETIC ANISOTROPIC COMPOSITE MATERIALS

The present invention concerns a new soft magnetic anisotropic composite material as well as a process for the preparation of this material.

The new composite material is characterized by high saturation flux density, high maximum permeability and low eddy current losses. These properties are the result of a considerably improved green density and indicate that the new composite material would be suitable for devices operating at power frequencies between 5.0 and 5000 Hz, e.g. relays, transformers, inductors and for magnetic shielding as well as for certain types of motors. The material can also be used for devices operating up to 50 kHz without significant eddy current losses.

In brief, the new high density composite material consists of compacted, flaky shaped iron particles bonded together by a non-magnetic organic resin, whereby the particles are aligned in an essentially parallel relationship. The high density, which in this context means a density above 7.4 g/cm^3, is mainly the result of the flaky form of the particles in combination with certain process steps such as the soft annealing step described below.

Materials of flaky shaped particles have previously been proposed for magnetic applications. Specifically, and contrary to the present invention, these materials are intended for static magnetic components such as magnetic cores. Thus the US patents 2 937 964 and 3 255 052 both concern magnetic cores made of flaky shaped particles of a nickel based alloy which also includes iron and molybdenum. According to the US patents the particles are insulated by a plurality of layers including i. a. silicate. The article "Compressed Iron Motor Core for Electric Motors" by Kiyoshi Fukui et al. in IEEE Transactions on Magnetics, September 1972, describes the use

of flaky electrolytic iron and spherical atomised iron powders of different sizes in compressed iron powder cores for small electric motors. The article "A laminated flake-iron powder material for use at audio and ultrasonic frequencies", Soft magnetic materials in Telecommunications, Pergamon Press, London 1953 pp 268 - 277 discloses an flaky shaped iron powder having a density of about 7.0 g/cm^3 which is taught not to be useful for power frequencies.

According to the present invention the new material is a soft magnetic, anisotropic composite material, which essentially consists of compacted, essentially flaky shaped, electrically insulated particles, which have been prepared by cold rolling and disintegration of an essentially pure iron powder. The particles are aligned in an essentially parallel relationship and bonded together by an organic polymer resin in an amount of 0.15 to 0.75 % by weight. The diameter of the particles is 3 to 35 times the thickness, preferably 5 to 20. A characterizing feature of the new material is the high density of at least 7.4 g/cm^3.

The present invention also concerns a process for the preparation of the composite material comprising the following steps:

a) cold rolling essentially pure iron powder into essentially flake shaped particles,
b) disintegration of the rolled powder to a maximum particle size of 500 micron
c) soft annealing the resulting powder at a temperature of 700 to 900 °C in a reducing atmosphere, such as H_2 atmosphere,

d) disintegrating of the annealed powder in order to obtain essentially the same particle size distribution as in step b)

e) mixing the powder with an organic binder resin,

f) feeding the obtained mixture into a pressing tool such that the flakes are aligned in the tool in a substantially parallel relationship,

g) compacting the material,

h) removing the compacted material from the pressing tool and, optionally,

i) stress relieving the material at an elevated temperature.

The starting material for the process is suitably an iron powder prepared by a conventional method, such as atomisation or direct reduction of iron ore particles. This powder is then annealed in order to reduce the content of impurities, such as carbon and oxygen, and to soften the iron. This operation is preferably carried out in a reducing atmosphere at a temperature of about 750-1000°C. The obtained powder contains less than 0.1 % by weight of carbon. Powders of this type are available from Höganäs AB, Sweden as ASC 100.29, which is an atomised powder containing less than 0.005 % by weight of carbon and NC 100.24 which is a sponge iron powder containing less than 0.01 % by weight of carbon. The oxygen contents are approximately 0.09 and 0.40% by weight, respectively. The annealed particles are then cold rolled into essentially flaky shape and disintegrated such that the diameter of the particles are 3 to 35 times the thickness and the maximum (diameter) particle size is about 500 µm. The flaky shaped particles thus obtained are then soft annealed at a temperature in the range of 700-900°C in a reducing e.g. H_2 atmosphere. In contrast to previously used soft annealing processes

in this field, the annealing process according to the invention is carried out at lower temperature and no inert inorganic powder material, such as aluminia, has to be added before the heating in order to prevent sintering. As a consequence no step for removing the inert material is included in the process according to the present invention. After the soft annealing step the carbon and oxygen contents of the annealed products are essentially the same as before this step. In order to secure the correct particle size distribution the annealed particles are subjected to an additional disintegration step. According to a preferred embodiment of the invention the iron flakes are then subjected to a phosphoric acid treatment in aqueous solution. The iron particles are subjected to the phosphoric acid at a temperature and for a time sufficient to form a thin electrically insulation layer around the individual iron flakes.

After the phosphoric acid treatment the powder is dried and mixed with an organic binder resin in an amount of less than 1 % by weight, preferably between 0.15 and 0.75% by weight and most preferably between 0.30 and 0.70 % by weight of the iron powder. If the binder content is less than 0.3 % the edge brittleness increases rapidly and makes the material hard to machine. The organic binder could be selected from thermosetting or thermoplastic resins and is preferably selected from the group consisting of epoxy resins such as Araldite, PPS (polyphenylene sulphide) or PEEK (polyetherether ketone).

The mixture of iron flakes and organic binder is then fed into a pressing tool such that the flakes are aligned in the tool in a substantially parallel relationship. This can be accomplished by allowing the flakes to fall freely into the die from a funnel which is positioned over the die, by vibrations, by magnetic

alignment or combinations thereof. The pressing tool could optionally be evacuated before the compaction of the flaky material, and, if the organic binder used is a thermoplastic resin, the material should be heated to a temperature above the melting point of the thermoplastic resin before the compacting step. The evacuation step is especially preferred if very high densities are required, and it has been found that va-cuum pressing increases the density by about 0.1 g/cm^3 which under certain circumstances is of great importance. Generally, the compacting step is carried out as a high-pressure isostatic or uniaxial pressing at pressures in the range of 400-1000 MPa. The compacting temperatures vary depending on the type of binder and the intended use of the final product. For epoxy resins the compacting step could e.g. be carried out at 70°C and a curing step might be carried out at 70-100°C. For PPS and PEEK type of resins the compacting could be carried out at 300°C and the crosslinking at 400-450°C. The compacting times are not critical but should be relatively short, such as 5-20 s, for economical reasons.

When removed from the pressing tool, the compacted material is either stress relieved at an elevated temperature or subjected to an elevated temperature and subsequently to a controlled cooling.

Due to the high densities, up to 7.58 g/cm^3, the properties of the new material are unique and similar to those of stacked 35-50 µm thick sheets of pure iron separated by very thin electric isolators. Thus, the bandwidth of the soft magnetic composite material can be as high as 100 kHz, the saturation flux density more than 1.9 Tesla and the maximum permeability, µmax. = 400. The mechanical properties of new material seem to have an optimum of about 150 MPa at a binder content of 0.35-0.50% by weight.

The invention is further illustrated by the following non-limiting example:

An atomised iron powder, ASC 100.29 (commercially available from Höganäs AB, Sweden) was used as base material for the new material according to the invention. The base powder consisted of irregularly, uniaxially shaped particles, which were rolled between two steel rolls in such a way that virtually each particle without contact with other particles was subjected to a press force corresponding to 3 ton/cm. After rolling the powder was disintegrated in order to separate particles which have stuck to each other during rolling, in order to obtain a powder having a maximum particle size of 420 µm. The obtained powder was in the form of flaky shaped particles having an average diameter of 250 µm and a thickness of 35 µm.

The powder was very hard as it has been subjected to strong deformation and, as a consequence, it was difficult to compact. The density when compacting at 800 MPa was 6.8 g/cm^3. The powder was soft annealed in a reducing H_2 atmosphere at 750°C during 45 minutes. At this temperature the iron powder could be soft annealed essentially without risking that the powder particles sintered together.

After the annealing step another disintegrating of the powder was carried out in order to restore its particle size distribution without deforming the particles which would once more result in hardening due to deformation. Bodies compacted with this powder had a density of 7.45 g/cm^3 (800 MPa), which can be compared with the density of the base material of 7.3 g/cm^3.

A thin insulating layer on the iron flakes was provided by subjecting the powder to a treatment with aqueous phosphoric acid. The oxygen and phosphorus contents of the obtained flakes were 0.41 and 0.02 % by weight, respectively.

The obtained powder was subsequently mixed with different amounts (from 0.2 to 1.0% by weight) of Araldite LY 5052, an epoxy resin available from Ciba-Geigy, and was compacted to ring cores for measuring of magnetic properties. After the compacting operation the ring cores were heated (80°C, 2 h), for curing of the epoxy binder. By compacting (800 MPa) the powder mixture in vacuum in an uniaxial tool a density of 7.58 g/cm^3 was obtained when the content of epoxy binder was 0.6% by weight. On average the vacuum compacting gave 0.1 g/cm^3 higher densities than conventional compacting in uniaxial tools. Densities of at least 7.4 g/cm^3 were observed for all components based on powders having an epoxy content between 0.2 and 0.7 also with conventional compacting.

A comparison between the results obtained with the material according to the present invention and a conventional material is given below.

Material	Density (g/cm^3)	Max. permeability	Total core loss at 0.5T; 400 Hz (W/kg)	Total core loss at 1.0T 400 Hz (W/kg)
Flakes+0.6% epoxy	7.58	390	20.9	85
ABM100.32 + 0.5% Kenolube 0.5% resin	7.18	225	35.0	140

CLAIMS

1. Soft magnetic, anisotropic composite material essentially consisting of compacted, essentially flaky shaped, electrically insulated particles of essentially pure iron powder containing less than 0.01 % by weight of carbon, which particles are aligned in essentially parallel relationship and bonded together by an organic polymer resin characterized by a green density of at least 7.4 g/cm^3 at a compaction pressure of 800 MPa.

2. Composite material according to claim 1, wherein the particles have a ratio diameter to thickness of 3 to 35 and an average thickness of 10-100 µm and an average diameter of 200-500 µm.

3. Composite material according to claim 1 or 2 wherein the organic polymer is present in an amount of less than 0.75, preferably between 0.30 and 0.70 % by weight of the iron powder.

4. Composite material according to any of the claims 1-3, wherein the electrically insulated flaky particles originate from atomised particles of essentially pure iron coated with an insulating layer.

5. Composite material according to any of the claims 1 - 4, characterized in that the organic polymer resin is a thermoplastic or thermosetting resin.

6. Composite material according to claim 5, characterized in that the organic polymer is an epoxy resin.

7. Process for the preparation of a soft magnetic, anisotropic composite material according to any of the preceding claims, characterized by the following steps:

a) cold rolling essentially pure iron powder into essentially flake shaped particles,

b) disintegration of the rolled powder to a maximum particle size of 500 micron

c) soft annealing the resulting powder at a temperature of 700 to 900 °C in a reducing atmosphere

d) disintegrating of the annealed powder in order to obtain essentially the same particle size distribution as in step b)

e) mixing the powder with an organic binder resin,

f) feeding the obtained mixture into a pressing tool such that the flakes are aligned in the tool in a substantially parallel relationship,

g) compacting the material,

h) removing the compacted material from the pressing tool and, optionally,

i) stress relieving the material at an elevated temperature.

8. Process according to claim 7 wherein the compacting step f) is carried out under reduced pressure or vacuum.

9. Process according to claim 7 or 8 wherein the particles of step d) are first subjected to a treatment with an aqueous solution of phosphoric acid at a temperature and for a time sufficient to form an insulating layer on the particles and subsequently dried.

10. Process according to claim 7 to 9 wherein the amount of binder is in the range of 0.15 to 0.75 % by weight of the iron powder.

11. Process according to any of the claims 7 to 10 wherein the binder is a thermosetting or thermoplastic resin.

12. Process according to claim 11 wherein the thermosetting resin is an epoxy resin.

13. Process according to claim 11 wherein the binder is a thermoplastic resin and the mixture in the pressing tool is subjected to a temperature above the melting point of the thermoplastic resin.

14. Process according to any of the claims 7 to 13 wherein the compacted material of step f) is subjected to an increased temperature and a subsequent controlled cooling.

15. Starting material for the preparation of soft magnetic, anisotropic composite material according to any of the claims 1-6, c h a r a c t e r i z e d in that it consists of flaky shaped particles of essentially pure iron containing less than 0.01 % by weight of carbon wherein the particles have a ratio diameter to thickness of 3 to 35, an average thickness of 10-100 µm and an average diameter of 200-500 µm optionally electrically insulated by an oxide layer.

16. Use of a composite material according to any of the claims 1 to 6 for devices operating at power frequencies, such as relays, transformers, inductors, motors and for magnetic shielding

17. Use according to claim 16 for devices operating between 50 and 5000 Hz.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/SE 96/00397

A. CLASSIFICATION OF SUBJECT MATTER

IPC6: B22F 3/02, C22C 33/02, H01F 1/24
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC6: B22F, H01F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

SE,DK,FI,NO classes as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

WPI

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	Soft Magnetic Materials in Telecommunications, Pergamon Press, London 1953, Campbell G et al: "A laminated flake iron powder material for use at audio and ultrasonic frequencies",page 268 - page 277, see page 268 - page 271 --	1-17
X	US 2937964 A (EDMOND ADAMS ET AL), 24 May 1960 (24.05.60), column 1, line 1 - column 2, line 16; column 5, line 23 - column 7, line 2 --	1-17
A	DE 3439397 C2 (VACUUMSCHMELZE GMBH), 18 January 1990 (18.01.90), page 2, line 1 - page 3, line 15; page 5, line 24 - line 31 --	1-17

[X] Further documents are listed in the continuation of Box C. [X] See patent family annex.

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INTERNATIONAL SEARCH REPORT

International application No.
PCT/SE 96/00397

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DE 3907090 A1 (VACUUMSCHMELZE GMBH), 6 Sept 1990 (06.09.90) --	1-17
A	US 4543208 A (H. HORIE ET AL), 24 Sept 1985 (24.09.85), column 3 - column 6 -- --------	1-17

INTERNATIONAL SEARCH REPORT

Information on patent family members

01/04/96

International application No.
PCT/SE 96/00397

Patent document cited in search report		Publication date	Patent family member(s)		Publication date
US-A-	2937964	24/05/60	NONE		
DE-C2-	3439397	18/01/90	NONE		
DE-A1-	3907090	06/09/90	NONE		
US-A-	4543208	24/09/85	EP-A,A,B	0112577	04/07/84
			SE-T3-	0112577	
			JP-B-	1051046	01/11/89
			JP-C-	1566650	25/06/90
			JP-A-	60016406	28/01/85
			JP-C-	1822409	10/02/94
			JP-B-	4016004	19/03/92
			JP-A-	59119710	11/07/84